Scope of Work and Schedule

Fate and Transport Modeling

Introduction

The U.S. EPA's October 12, 2012 letter (USEPA, 2012) requested that the Respondents perform fate and transport modeling at the West Lake Landfill (the Site). This Scope of Work (SOW) describes the modeling approach proposed to estimate potential fluxes of landfill leachate, possible radionuclide concentrations within the leachate, and the potential for transport of any radionuclide-contaminated leachate within the subsurface.

This SOW first outlines the objectives of the proposed modeling task. This is followed by a discussion of the general conceptual site model (CSM). Features of the Site that are expected to be simulated are described together with potential events and the physical and chemical transport processes that are envisioned as being incorporated in the modeling analyses. After describing the CSM and defining the objectives of the modeling calculations - which together define the necessary capabilities of the developed model – the calculation approach and the simulation programs proposed to meet the modeling objectives are identified. The final suite of scenarios to be simulated will be determined as part of the model implementation task.

It is assumed that modeling calculations will be performed on the basis of existing site-specific data, augmented where necessary with information and values obtained from technical literature and/or derived from professional experience.

Background

West Lake Landfill is located within the western portion of the St. Louis metropolitan area approximately two miles east of the Missouri River. Two areas of the Site contain radionuclides as a result of the use of soils mixed with leached barium sulfate residue as cover for municipal refuse. The Site is divided into two Operable Units (OUs). OU-1 consists of the two areas within the landfill where radionuclides are present and the area formerly described as the Ford Property, now called the Buffer Zone/Crossroad Property. OU-2 consists of other landfill areas that are not impacted by radionuclides (USEPA, 2008). Modeling calculations proposed in this SOW address the potential fate of radionuclides within OU-1. The nature and extent of radionuclides within OU-1 are discussed in several documents included in the administrative record for this site, including the Remedial Investigation (EMSI, 2000) and a Supplemental Feasibility Study (SFS) (EMSI, 2011) for OU-1.

~~The selected remedy for OU-1 presented in the Record of Decision (ROD) includes source control through containment of waste materials and institutional controls for the landfilled waste materials (USEPA, 2008). Components of the ROD-selected remedy include the following:~~

Commented [cao1]: Not sure what this letter is based on as far as Board recommendations is concerned since no final Board memo had been sent by this date --

Commented [cao2]: Board's draft recommendations prepared between February and May 2012 recommended gathering additional data to …

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Commented [cao3]: In several places, language appears inconsistent with Board's spring 2012 draft recommendations. For example, "within the landfill" together with "as cover for municipal refuse" in the previous sentence, and statements made on page 4 below, seems inconsistent with the Board's initial observations/comments/recommendations contained which included the following statements: 1) "The Board notes that the 1982 NRC Radiological Survey states that the representation of subsurface contamination based on auger hole measurements in Figures 15 – 19 of that report "are consistent with the operating history of the site, which suggests that the contaminated material was moved onto the site within a few days' time and spread as cover over fill material. Thus, one would expect a fairly continuous, thin layer of contamination, as indicated by survey results." (p. 16). The Board also notes that the most intense gamma peak readings for RIM in Area 2 are located within three feet of the surface (e.g., PVC 7, PVC-10, PVC-11); see Table 6-9 of RI report." 2) "The Board notes that Table 6-8 in the RI indicates that the estimated average total thickness of RIM for Area 1 is 3.37 ft, and 3.73 for Area 2; this is further supported by Table 5 attached to the 1982 NRC report. The RI report also indicates that "Based upon the radiological data, McLaren/Hart concluded that the zone of radiological impacts in Area 1 is generally a thin layer (5-feet thick or less) in the upper part of the landfill debris" (page 32) and "Based upon the radiological data, McLaren/Hart concluded that the zone of radiological impact in Area 2 is generally a thin layer (less than 5 feet) in the upper part of the landfill debris" (page 33). This conclusion is similar to the one made by the NRC in its 1982 Radiological Survey that the deposits appear to form "a fairly continuous, thin layer of contamination, as indicated by survey results (page 16) and "a contiguous layer" (page 21), reflected also in Figures 10 – 19 attached to that report which include a number of cross-section diagrams." 3) "Also, the Board notes that the RI report states that "Based upon the results of the downhole gamma logging and the laboratory analyses, radiologically impacted materials were generally found at depths ranging between 0 to approximately 6 feet in the northern portion of Area 2" and "In the southern part of Area 2, radiologically impacted materials were identified at depths generally ranging between 0 and 6 feet." (RI page 97)."

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Commented [cao4]: In light of Board's 2012 review (plus original reasons for doing the SFS), this next paragraph seems out of place/confusing/potentially misleading,

1. ~~A new landfill cover over the existing surface of Areas 1 and 2;~~
2. ~~Consolidation of radiologically contaminated surface soil from the Buffer Zone/Crossroad Property to the containment area;~~
3. ~~Groundwater monitoring and protection standards consistent with requirements for uranium mill tailing sites and sanitary landfills;~~
4. ~~Surface water runoff control;~~
5. ~~Gas monitoring and control including radon and decomposition gas as necessary;~~
6. ~~Institutional controls; and~~
7. ~~Long-term surveillance and maintenance of the remedy.~~

A ROD was signed in 2008. In addition, an SFS done in 2011 discussed potentially appropriate ~~P~~performance standards for cleanup of this site. ~~for these remedy components are detailed in the ROD. The following additional performance standards were also identified for the OU-1 remedy~~ (EMSI, 2011):

- ~~The proposed~~ A cap that ~~should~~ would meet the Uranium Mill Tailings Radiation Control Act of 1978 (UMTRCA) guidance for a 1,000-year design period including an additional thickness to prevent radiation emissions;
- ~~Air monitoring stations for radioactive materials should be installed at both on-site and off-site locations;~~
- Groundwater monitoring that would ~~should~~ be implemented at the waste management unit boundary and at off-site locations; and
- Flood control measures at the Site that would ~~should~~ meet or exceed design standards for a 500-year storm event under the assumption that the existing levee system is breached.

~~As defined in the OU-1 ROD, the new landfill cover for Areas 1 and 2 would consist of the following, from bottom to top: 2-ft of rock consisting of well-graded pit run rock and/or concrete/asphaltic rubble ranging from sand-sized up to 8-inches; 2-ft of compacted clay or silt that when compacted at optimum moisture content possesses a coefficient of permeability of 1 x 10^-5^ cm/sec or less; and 1-ft of soil suitable of supporting vegetative growth. These layer thicknesses are based on requirements of the Missouri Solid Waste Rules and the description of the cover system in the ROD (USEPA, 2008).~~ [A separate task will evaluate potential alternative landfill cover designs other than those discussed in the 2008 ROD, including possible use of an Evapotranspiration (ET) cover or ~~incorporation of a~~ geomembrane ~~into the design of the ROD-selected landfill~~ cover.]

Modeling Objectives

The proposed fate-and-transport modeling will provide site-specific calculations of the potential for radionuclides to leach from the landfill, reach the underlying saturated aquifer, and result in unacceptable concentrations within groundwater or surface water downgradient of the landfill. The following modeling objectives are proposed:

1. Calculate the potential for migration of leachate containing radionuclides from waste materials:
   a. Under current conditions, to validate the modeling approach and potentially bound parameter values for later predictive analyses;

Commented [cao5]: Board has said in its memos that monitoring by itself is not a CERCLA remedy (so could not be a "component of the ROD selected remedy"

Commented [cao6]: Not sure what this means --

Commented [cao7]: This makes it sound like they have been selected as part of the remedy – ROD would do that, not SFS (if the EMSI reference is to SFS)

Commented [cao8]: If this is the SFS, should identify it as such

Commented [cao9]: Not sure what this "proposed" refers to --

Commented [cao10]: How is this relevant to this SOW for ground water? Board recommendations on ground water don't address air do they?

Commented [cao11]: Again, as Board has stated, monitoring by itself is not a remedy, so not clear what performance standards (as discussed in the NCP) would be for here --

Commented [cao12]: The Board's draft recommendations from spring of 2012 questioned use of state's subtitle D regs --

Commented [cao13]: See comment 4

Commented [cao14]: Should explain why this might be relevant to ground water SOW --

Commented [cao15]: The Board's draft recommendations from spring of 2012 included other things that are relevant to this SOW for ground water, including language from the initial draft ("Groundwater: monitoring wells placed in perimeter fashion; dated GW data—gather new data now; wells seem to be clustered—large gaps—need wells in between gaps to determine if there is, in fact, a plume issue (e.g., predesign installation of new wells); if we can't fully characterize GW, then we need to have a sufficient record to substantiate that conclusion"), as well as later versions which said: "Based on the information presented to the Board, it appears that there have been some samples of groundwater at this site that exceed standards considered as ARARs. The Region also stated that no discernable plume at this site has been identified, and its preferred approach is to continue monitoring groundwater. Generally, under existing Agency guidance, exceeding a maximum contaminant level in groundwater normally would warrant a response action (OSWER Directive 9355.0-30, *Role of the Baseline Risk Assessment in Superfund Remedy Selection* Decisions and OSWER Directive 9283.1-33 *Summary of Key Existing EPA CERCLA Policies for Groundwater* Restoration). The Board recommends that the Region consider additional wells at the site to better delineate the vertical and lateral extent of potential site-related contamination previously indentified from limited sampling in Area 1 and especially Area 2. These additional wells would be instrumental in clarifying the presence of an isolated groundwater hot-spot versus a groundwater plume in the complex subsurface geologic setting. In addition, the Region should explain why there are numerous decommissioned wells on site. Sampling of these wells may have provided a more complete picture of potential groundwater contamination. The general recommendation is that the additional wells be nested along the western border (Crossroad property) of Area 2 in the unconsolidated alluvial deposits and the underlying fractured and vuggy, limestone Keokuk formation. The Board also notes that the Agency's long-standing policy has been that monitoring by itself is not a CERCLA remedial action, and believes that the information submitted to the Board may not support a conclusion that monitoring to evaluate effectiveness of the source control remedy (if that approach is selected) would constitute an effective or final groundwater response action for this site. As such, the Board recommends that the decision documents clearly expl ...

b. Under future conditions, assuming the emplacement of a new landfill cover for OU-1; and
c. Under future conditions, following the period of active maintenance of the new landfill cover for OU-1.

2. Calculate the potential for leachate containing radionuclides to migrate vertically through waste that is uncontaminated by radiological constituents and through native materials beneath the landfill, and to impact underlying groundwater;

If the prior calculations indicate that a potentially measurable impact to groundwater may occur:

3. Calculate the likely fate of any radionuclides that reach groundwater, and the potential for the development of a contaminant plume;
4. Calculate concentrations over time of radionuclides in groundwater at defined locations including, but not limited to, the property fence line/boundary; and
5. Evaluate the potential for radionuclides that reach the groundwater to migrate toward, and discharge to, the Missouri River at levels exceeding standards.

These are the specific objectives of the proposed modeling task. The model may, at some later time, be used to support other Site objectives such as (a) designing a suitable groundwater monitoring program, including defining the locations and frequency of sampling to detect any potential off-site migration of radionuclide constituents and/or (b) evaluating alternative landfill cover designs such as an ET cover or incorporation of a geomembrane.

Fate and Transport Conceptual Site Model

Because the overall mass of radium at the Site is small[1] and future infiltration through the landfill materials will be less than at present due to the planned emplacement of an additional landfill cover over the existing landfill cover material, it might be expected that concentrations of radium will necessarily decline in the future. However, site-specific conditions need to be evaluated before reaching this conclusion. For example, uranium and thorium that are present in the waste materials will continue to decay, and in doing so, generate radium. In addition, the landfill and groundwater geochemistry will change over time due to the eventual exhaustion of readily-biodegradable organic matter in the landfill. This will in turn affect the stability of some minerals available to sequester radium.

Selection of an appropriate calculation method, and of a suitable simulation code or suite of codes to implement the calculations, requires that the modeling requirements are defined. In the context of radionuclides, the Nuclear Energy Agency Organization for Economic Co-operation and Development (NEA-OECD, 2000) developed a systematic approach to define relevant scenarios for safety assessment studies at radioactive waste management sites. This consists of identifying and prioritizing the Features,

[1] Using the arithmetic mean concentrations (reported as pCi/gram) from Appendix A of the RI, as well as an estimated mass of soils for the Area 1 and 2 surface and subsurface zones at the West Lake site, a preliminary estimate of the amount of ^{226}Ra at the site indicates that there is less than 40 grams of ^{226}Ra within Areas 1 and 2.

Commented [cao16]: This seems to suggest that it's mass that counts, not risk posed by the rad – based on discussions during the February, 2012 Board meeting and draft recommendations prepared by the Board during the spring of 2012, this statement/approach appears inconsistent with the documents in the administrative record for this site and with the remedial program's approach at rad sites around the country. For example, the Board's draft recommendations stated:
"Finally, the Board notes that the FS (at page 60) stated that "Excavation of a smaller volume of radioactively impacted material [than the estimated 250,000 cubic yards of total RIM plus soil and debris] would not significantly reduce the threat posed by the overall site." The Board is concerned that this kind of approach is inconsistent with, and could undermine, ongoing cleanup of rad sites in several other Regions. Specifically, in Region 2, reduction of rad-impacted source material is being undertaken in a manner that is protective and without short-term impacts, where the Region determined that eliminating the source is an important objective of the cleanup. Region 2 has been removing radiological contamination from residential and commercial properties for the past two decades. That work is undertaken with appropriate engineering controls and in accordance with approved health and safety plans, often with homeowners remaining in their residences during the cleanup effort. These types of cleanups can be safely and efficiently undertaken. Given the presence of highly radioactive material at this site, and the fact that its hazardous nature will continue to increase over time, the Board believes excavating and/or treating any amount of the RIM should lead to important risk reduction. Where it appears that much if not all of the RIM is located near the surface, cleanup at this site appears less complicated than other sites where, for example, buried drums containing liquids have been safely excavated. Radiological material is also easily sorted out in the field with portable instruments that provide instantaneous measurements to ensure that only contaminated material is retrieved which, in turn, minimizes disposal costs."
Saying that "the overall mass of radium at the Site" also could cause confusion/misunderstanding

Commented [cao17]: This is making an assumption about the remedy that doesn't seem to take into account the Board's concerns/ recommendations made in draft memos during spring of 2012 – the Board didn't make its recommendations regarding groundwater based on an additional landfill cover; rather, the recommendations were that there's insufficient data and more data/more wells are needed to adequately characterize the site (see comment 15 for actual wording of Board's spring 2012 1draft recommendations/comments)

Commented [cao18]: This appears inconsistent with Board's views expressed during meeting and in spring 2012 draft recommendations – for example: "Based on the package provided to the Board, it appears that there are potentially significant amounts of RIM that are highly toxic (e.g., based on NRC estimates in the 1982 and 1988 reports, radium of up to 22,000 pCi/gr, bismuth-214 of up to 19,000 pCi/g, and average thorium-230 concentrations of 9000 pCi/gr; the package at page 44 notes that the RI report discussed thorium-230 at levels as high as 57,300 pCi/gr) and that the highest gamma peak intensity readings are at shallow depths. The FS states (page 84) that most of Area 2 contains RIM at above 100 pCi/gr. The NRC reports also discuss h ...

Commented [cao19]: See comment 17 --

Commented [cao20]: Consistent with Board comments/recommendations made at other rad sites (e.g., Hanford), CERCLA remedy selection should be done using CERCLA rad guidance --

Events, and Processes (*FEPs*[2]) that potentially affect the fate and transport of radionuclides at a site, and developing and modeling individual scenarios, each of which consists of a well-defined, connected sequence of selected FEPs. This SOW identifies principal FEPs for the Site that it is anticipated will require consideration in the modeling analyses. However, the final site-specific FEPs and the suite of simulation scenarios will be defined during the implementation phase of the modeling task.

Primary Site-Specific Features

An overview of the primary features that affect radionuclide fate and transport is provided here. The source of radionuclides of potential concern is leached barium sulfate residue mixed with soil ~~and used as daily and intermediate cover for municipal solid waste deposited in landfill~~ in Areas 1 and 2. ~~This radiologically-impacted material (RIM) is currently covered by old landfill cover material~~. Underlying the RIM is refuse that does not contain radionuclides, and under that is partially saturated alluvium. Over time some fraction of radionuclide-bearing water could potentially percolate vertically to reach the water table. According to the RI [EMSI, 2000], the saturated aquifer largely consists of alluvial sand, underlain by more impervious limestone and dolomite bedrock. The horizontal hydraulic gradient within the aquifer is relatively flat, which would tend to result in slow advection along a trajectory that intersects the Missouri River downgradient of the Site. If radionuclide-containing water currently located within or under OU-1 were to reach the water table beneath the landfill, then mixing, dispersion, and dilution of that radionuclide-containing water would occur at the water table beneath the landfill, within the saturated aquifer, and within the hyporheic zone of the Missouri River.

A dominant feature [which, depending upon the simulation scenario, may also constitute an event] that must be considered in the modeling calculations, and for which a design is presented in the ROD but for which potential alternatives have since been identified by USEPA for evaluation, is the new landfill cover to be installed over the current surface of the old landfill cover. Modeling calculations proposed under this SOW will only consider the ROD-selected landfill cover, the design of which is detailed above and within the ROD (USEPA, 2008). However, the developed model could be used at some later time to evaluate alternative cover designs such as an ET cap and/or the incorporation of a geomembrane within the ROD-selected landfill cover.

Primary Site-Specific Events

Several events may affect the landfill water balance, the potential for radionuclide partitioning and migration, and the potential for radionuclide transport within the partially saturated and saturated zones at the Site. Example events are summarized in Table 1.

Commented [cao21]: See comment 3 above --

Commented [cao22]: See Board's spring 2012 draft recommendations/comments describing documents in the administrative record that describe RIM as at the surface in certain locations, and between 0 – 6 feet in many others.

Commented [cao23]: Should it be "and" or should it be "or" ?

Commented [cao24]: This makes it sounds like a certainty – Ron indicated at the Board meeting and other conversations that it might not be so – for example, the fact that Kaarst is a factor, plus the hits above MCL that have been documented in the administrative record might not support this --

Commented [cao25]: See comment 17 above

Commented [cao26]: See comment 4 above – this approach appears to ignore Board review process and the spring 2012 draft recommendations/comments

[2] The following definitions apply (Sandia National Laboratories, 2010):
Feature – An object, structure, or condition that has a potential to affect repository system performance.
Event – A natural or human-caused phenomenon that has a potential to affect repository system performance and that occurs during an interval that is short compared to the period of performance.
Process – A natural or human-caused phenomenon that has a potential to affect repository system performance and that occurs during all or a significant part of the period of performance.

Table 1 Primary Events and Processes of Potential Radionuclide Fate and Transport at the Site.

FEP Element	Description
Events:	1. Transition from current cover conditions to final cover under active maintenance: • Cover design (*2-ft of well-graded pit run rock and/or concrete/asphaltic rubble; 2-ft of compacted clay or silt with a coefficient of permeability of 1 x* 10^{-5} *cm/sec or less; and 1-ft of soil suitable of supporting vegetative growth*) • Period of active maintenance (*30 yr min/200 yr ROD/1000 yr UMTRA-compliant*) 2. Transition from active maintenance period to post-active maintenance period: • Intermediate infiltration rates (*reduced by grade, vegetation, etc.*) 3. [Bio-]degradation of landfill wastes: • Degradation time-frame (*rapid versus extended time*) • Effects and duration on chemistry (*oxidation-reduction* [*redox*], *carbonate, CO2, pH, etc.*) 4. Flood events: • 500 year
Processes:	1. Net infiltration: • Under current conditions • During period of active cover maintenance (*incorporating ET as a process*) • Following period of active cover maintenance (*reduced by grade, vegetation, etc.*) 2. Ingrowth of radium from uranium and thorium decay: 3. Partitioning of radium, uranium, thorium from soil to water/landfill leachate: • Decay/ingrowth, sorption/complexation, mineral dissolution/precipitation 4. Transport within the partially-saturated zone: 5. Mixing at the water table: • Depth of penetration, and dilution • Sorption/complexation, mineral dissolution/precipitation 6. Transport within the saturated aquifer: • Advection, dispersion, decay/ingrowth, sorption/complexation, mineral dissolution/precipitation 7. Discharge to, and mixing with, Missouri River: • Hyporheic zone chemical process • Sorption/complexation, mineral dissolution/precipitation

The Uranium Mill Tailings Remediation Program (UMTRA) focused on the design of purpose-built repositories for uranium tailings piles; however, the UMTRA containment design time-frame of 1000 years is a guide for other radionuclide wastes.

One important event is the grading of Areas 1 and 2 and the emplacement of the final landfill cover on top of the current landfill cover material in these areas. This new cover will greatly reduce infiltration and the potential for mass transfer of radionuclides to mobile water for the period of active maintenance. If active maintenance were to cease, over some time the effectiveness of the landfill cover may decline, potentially resulting in an increased infiltration rate. However, infiltration rates following cessation of active cover maintenance would be expected to be lower than under current

Commented [cao27]: See comments 17 and 25

Commented [cao28]: ditto

Commented [cao29]: Does the period of active maintenance here equal 1000Years?

See Board comments that: "However, the package lacked sufficient information on the long term protectiveness of the preferred remedy. Specifically, how the preferred remedy remains protective given the increasing daughter ingrowth concentrations of radium 226/228, radon 222, and the increase in toxicity over time (1000 years).

Both of these landfill designs (Subtitle D and UMTRCA), as in the preferred alternative, have shortcomings for RIM waste itself and especially in a humid region. A comparison of various landfill capping designs addressing both humid region conditions and long term protection from RIM (1000 years) would be an important concept for the preferred remedy. However, the package did not appear to include alternative cap designs, i.e., EPA landfill cap guidance design, existing cap designs for similar RIM at Weldon Springs, or evapotranspiration cover cap system designs (OSWER Fact Sheets: EPA 5420F-03-015, 2003; EPA 542-F11-001, 2011). For example; a Subtitled C/UMTRCA hybrid may be suitable for both long term infiltration management and radiation shielding protection, The Board recommends that the region include in its remedy selection process evaluations of cap designs similar, but not limited to, the above conditions and guidances. The package also does not address several aspects of the potential for future migration of contamination to ground water. The current lack of a discernable plume above MCL levels may not be a sufficient basis to determine there is little or no potential for there ever to be one. Particularly in light of the long-lived toxic nature of the radioactive contaminants as well as chemical and physical changes over time at the landfill, the Board recommends that a more rigorous evaluation of potential migration to groundwater be undertaken. The evaluation should not assume that pumping at the former active sanitary landfill will continue, unless that is part of this remedy. For these reasons, the Board recommends that the region provide further information on alternative cap designs plus fate and transport of groundwater that supports the preferred remedy's long term protectiveness.

conditions since the cover design incorporates a grade (whereas, the majority of the current landfill cover is flat) and other features that would endure for many years following cessation of active maintenance.

Another important event is the slowing rate of biodegradation of organic materials in the landfill over time; this will alter the geochemistry within the landfill wastes and impact radionuclide partitioning between mobile and immobile phases in the refuse that contains RIM, the underlying refuse that does not contain RIM, and potentially the underlying alluvial aquifer.

Primary Site-Specific Processes

Several processes may affect the potential for radionuclide partitioning and migration, and the potential for radionuclide transport within the partially saturated and saturated zones at the Site. Example processes are summarized in Table 1. One important process is the complex interaction of the RIM with the surrounding pore water, and the role of pore water and soil chemistry on the potential for radionuclide partitioning and migration. Since radionuclide geochemistry will be an important process in the modeling scenarios, an overview of relevant radionuclide geochemistry is provided below.

Geochemistry of Radionuclide Decay, Ingrowth, Partitioning and Migration

Radium Geochemistry

Radium dominantly occurs within leached barium sulfate residues that were mixed with soil ~~and used as daily and intermediate soil cover for solid waste disposed at~~ Areas 1 and 2. The co-precipitation of radium into barium sulfate is a well known process to control radium (Doerner and Hoskins, 1925; Bruno et al., 2007; Zhu 2004a, 2004b; Mahoney 1998, 2001; Grandia et al., 2008; Bosbach et al., 2010). Consequently, equilibrium between pore water and the radium component of barium sulfate will define the initial radium source term leached from the RIM.

Radium may also be attenuated in clean alluvium and groundwater via adsorption onto iron-bearing minerals, ion exchange on clays, and co-precipitation with other sulfate and carbonate minerals such as gypsum and calcite. Of these mechanisms, co-precipitation is expected to be the dominant process close to the landfill due to the sandy nature of the aquifer and expectedly low redox conditions (making iron oxyhydroxides unstable). Downgradient of the landfill - and increasingly within the landfill over time - more oxidizing conditions may be present, and the abundance of iron-bearing minerals available for radium adsorption may increase. Another important consequence of the change in landfill biogeochemistry over time is the likely increase in pH as readily-biodegradable material is consumed. As pH increases, the amount of calcite that will precipitate will increase, and radium co-precipitation with calcite will be more favored, reducing its mobility.

Uranium Geochemistry

Uranium and thorium are important because they occur within the RIM and they decay over time, producing additional radium. Under current conditions uranium concentrations are expected to be controlled by uraninite (UO_2) due to the reducing conditions within the landfill. If oxidizing conditions

Commented [cao30]: How does this take into account the ingrowth issue (the RIM will getter hotter over time) that the Board identified/discussed during review meeting and in spring 2012 draft recommendations?

Commented [cao31]: How much of this is there – see comment 3 above, where Board discussed various documents in the administrative record indicating that "Thus, one would expect a fairly continuous, thin layer of contamination, as indicated by survey results."

Commented [cao32]: See above comments (e.g., #3)

Commented [cao33]: Is this expectation discussed/supported in the administrative record (FS? SFS?)

Commented [cao34]: ditto

Commented [cao35]: see comment 31

Commented [cao36]: in light of Board's spring 2012 draft recommendations/comments (see #18 and #22 above), is the "likely increase" here explained in the FS or SFS?

Commented [cao37]: See comment 33 above

Commented [cao38]: Is this expectation affected by Board's spring 2012 draft recommendations/comments?

return, however, then uranium solubility could be controlled by the generally more soluble U^{+6} (uranyl) minerals such as schoepite [$UO_2(OH)_2 \cdot 2H_2O$] or less soluble forms such as carnotite (KUO_2VO_4) and tyuyamunite [$Ca(UO_2)_2(VO_4)_2$] (Tokunaga et al., 2009). In addition to the oxidation state of uranium, other factors affecting dissolved concentrations include levels of dissolved carbonate generated by biodegradation (which increase solubility) and presence of iron oxyhydroxides (which decrease solubility).

Thorium Geochemistry

Thorium is not redox sensitive and solubility conditions will be controlled by thorianite (ThO_2) under all redox conditions. Complexation reactions that form thorium carbonate complexes are not as significant as those for uranyl carbonate complexes, but they will play a role in thorianite solubility calculations. Reductions in carbon dioxide pressures will also reduce thorium concentrations in groundwater.

The long-term in-growth of ^{226}Ra from ^{230}Th is complicated by the fact that the majority of in-growth radium will be retained within the crystal structure of the thorianite (ThO_2). Only a small fraction of the radium that is produced from the decay of thorium will have the potential to be released to groundwater. This fraction is expected to be derived from near the surface of the thorianite crystals.

Commented [cao39]: See comments 30 – 38 on previous page – in light of Board's spring 2012 draft recommendations/comments, is this conclusion specifically discussed/supported in the FS or SFS?

Calculation Approach

General

The approach to undertaking modeling calculations will follow the sequence of steps defined below:

- Determine and document final FEPs;
- Identify simulation scenarios, based on the final FEPs;
- Identify parameter ranges and uncertainties;
- Develop necessary model(s);
- Complete model calculations; and
- Present and interpret results.

As the modeling is implemented, there will be some iteration between steps in the sequence. It is expected that there will be communication and interaction with USEPA to seek input on the FEPs, simulation scenarios, and parameter ranges and uncertainties identified for inclusion in the modeling prior to undertaking the model calculations. It is envisioned that communication and interaction will include the following:

- Presentation and discussion of certain detailed or fundamental concepts – such as the CSM, FEPs and scenarios for inclusion in the modeling;
- Discussion of other less critical aspects of the modeling task; and
- Presentation of intermediate deliverables for review and discussion.

Graded Approach

A graded approach is proposed to undertake the modeling analyses (USEPA 2002, 2009). This graded approach will:

- Use relatively simple methods for initial calculations under the premise that the inherent conservatisms are protective of groundwater and other receptors. Increasing simulation rigor will only be used, if necessary, if simpler approach(es) yield unreasonable results.
- Provide a mechanism to cease model calculations if it becomes evident that no further calculations are necessary. For example, saturated zone flow and transport calculations will only be undertaken if geochemical and vadose zone modeling calculations suggest that a potentially measurable impact to groundwater could occur.

The modeling approach and specific model calculations will be designed to incorporate the principal FEPs while mitigating the potential for computationally-intensive calculations that prevent a thorough exploration of parameter variability and scenario uncertainty. Multiple scenarios will be simulated to evaluate the potential impact of scenario uncertainty on model outcomes, while sensitivity analysis will be used to evaluate the potential impact of parameter variability on model outcomes.

Modeling analyses will be designed to predict the concentration of radium in groundwater for a period of 1,000 years. Concentrations will be forecast at defined compliance locations including, but not limited to, the property fence line/boundary, for the 1,000-year period and will be compared to regulatory standards. If regulatory standards are not exceeded then no further analyses will be required. However, if simulated concentrations exceed regulatory standards, the graded approach will be used to identify the technical element of the modeling approach that incurs the most inherent conservatism in the calculations so that element of the modeling approach can be treated more rigorously to reduce that inherent conservatism (Dixon et al, 2008). If the graded simulation approach has been applied until all inherent conservatisms have been reduced or eliminated, yet simulated concentrations exceed regulatory standards, then this will be considered to be a reliable result.

Commented [cao40]: The administrative record already includes well samples that show MCLs are exceeded. See comment 15 above.

Simulation Code Selection

Table 1 outlines primary events and processes that will be considered in the calculations. The range of potential outcomes will be evaluated by performing several model simulations that consider reasonable alternate conceptualizations of subsurface conditions. Since parameterization of the geochemical component of the model is likely subject to more variability and uncertainty than the groundwater flow component of the model - given the large number of chemical processes that potentially affect radium fate and transport – advective-dispersive migration will be simulated as one-dimensional (1-D), coupled with a rigorous treatment of the complex geochemical processes. The following sequential series of calculations is proposed to collectively comprise the model [consistent with the graded approach, some calculations will only be undertaken if necessary based on the results of preceding calculations]:

1. The Hydrologic Evaluation of Landfill Performance (HELP) code will be used to determine the run-off component of the surface-water balance and remaining water available for infiltration through cover materials under current conditions, final cover conditions, and following the period of active cover maintenance;
2. HYDRUS 1-D (Simunek et al., 1998) will be used to simulate the water balance in the subsurface (after run-off has been accounted for) and the migration of infiltrating water;
3. The USGS-supported geochemical simulation software, PHREEQC (Parkhurst and Appelo, 1999), which is linked to HYDRUS through the HP1 program (Jacques and Simunek, 2005), will be executed simultaneously to provide concentrations of radionuclides in the leachate as it moves within the unsaturated refuse and underlying unsaturated alluvium;
4. The depth of penetration of any leachate that reaches the water table will be calculated using an established method such as that detailed by USEPA (1996);
5. PHREEQC, linked with HYDRUS, will then be used to calculate the effects of mixing on geochemistry that occurs between the leachate and groundwater at the water table;
6. Output from these calculations will provide the time-varying groundwater composition for simulating 1-D radionuclide fate and transport within the saturated zone toward the Missouri River using PHREEQC; and
7. PHREEQC will be used to represent geochemical processes that may occur within the hyporheic zone of the Missouri River.

Commented [cao41]: See comments above (e.g., #4, #17 etc)

<u>Overview of HELP Calculations</u>

HELP (Schroeder, P.R. et al, 1994a, 1994b; Berger, 2011; Berger and Schroeder, 2012) is a program originally developed by USEPA to evaluate the effectiveness of landfill cover designs. HELP will first be used to estimate the typical, quasi-steady-state surface-water balance, emphasizing the run-off rate and the net water available for infiltration through the current landfill cover. The purpose of these calculations is solely to support validation of the modeling approach and constrain the values of certain parameters to be consistent with historical water samples. HELP will then be used to make similar calculations to estimate run-off and the net water available for infiltration through the new landfill cover that would be constructed under the ROD-selected remedy, and to estimate run-off and the net water available for infiltration through the new cover following the period of active maintenance. Alternate periods of active maintenance may be considered in alternate simulation scenarios. The HELP model can explicitly account for rainfall-runoff under alternate cover designs, including alternate slopes (grades).

Commented [cao42]: See comments above (e.g., #4, #17 etc)

<u>Overview of HYDRUS 1-D Calculations</u>

HYDRUS-1D (Simunek et al., 1998) is a public domain Windows-based modeling environment that simulates the movement of water, heat, and multiple solutes in variably saturated media. The flow equation formulation in HYDRUS incorporates a sink term to account for water uptake by plant roots, as well as a dual-porosity type flow capability in which one fraction of the water content is mobile and another fraction is immobile. The solute transport equations consider advective-dispersive transport in the liquid phase, as well as diffusion in the gaseous phase. HYDRUS 1-D (Simunek et al., 1998) will be used to simulate the water balance in the subsurface (after run-off has been accounted for), and the migration of infiltrating water.

HYDRUS 1-D is linked to PHREEQC through the HP1 modeling software (Jacques and Simunek, 2005). This allows simulation of complex bio-geochemical reactions. Consistent with the graded modeling approach, the initial simulations will assume that radionuclide attenuation in landfill leachate only occurs in groundwater. However, the HP1 software may be used to estimate attenuation in the non-radiologically impacted refuse and unsaturated alluvium underlying Areas 1 and 2 if unreasonable results are obtained using the more conservative simplifying assumption.

Overview of PHREEQC Calculations

Geochemical modeling will first be completed to estimate the leaching potential of various radionuclides under current site conditions. The purpose of these calculations is to support validation of the groundwater modeling approach and constrain the values of certain parameters to be consistent with historical water samples. Following these calculations, the modeling will be used to evaluate the leaching potential under long-term future conditions under the ROD-selected remedy.

Commented [cao43]: ditto

Geochemical modeling methods to estimate source term concentrations for the radio-isotopes will primarily rely upon equilibrium thermodynamics and will be based upon mineral solubility relationships using current ground water compositions. Calculations will be performed using PHREEQC (Parkhurst and Apello, 1999). Solubility calculations for end member phases will be used for thorium and uranium. Radium will be assumed to be present as a solid-solution in barite with a lower thermodynamic activity. Solubility constants for uranium and thorium will, for the most part, be based upon the OECD NEA compilations (Guillaumont et al., 2003; and Rand et al., 2008). Other data sources will be used as needed (Dong and Brooks, 2006, 2008; Duro et al., 2006; Langmuir, 1978; Tokunaga et al., 2009). The ingrowth of ^{226}Ra from ^{230}Th is a time dependent process and the kinetics capabilities in PHREEQC will be used to estimate the production of ^{226}Ra for a period of 1,000 years.

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1-D transport modeling will also be performed with PHREEQC. Modeling will simulate a chemical system that is sufficiently complex to include the effects of landfill and groundwater geochemistry described above. Site-specific groundwater and soil data for uranium, thorium, and radium will define initial concentrations for these isotopes. The site analytical results, particularly the groundwater analyses, will also provide details on the overall geochemical environment of the landfill. The PHREEQC fate and transport model will include the following features:

- The effect of radium in-growth from the decay of thorium over time;
- Decreased methane generation and a possible change in site redox conditions from the reducing conditions currently present at the site to more oxidizing conditions;
- Radionuclide precipitation and/or co-precipitation, such as the partitioning of radium into calcite (Yoshida et al., 2008) present within the landfill;
- Changes in iron stability and potential precipitation of iron-bearing phases for the adsorption of radionuclides; and

- Adsorption reactions (surface complexation and ion exchange) (Dzombak and Morel, 1990; Mahoney et al. 2009a, b; Rojo, et al., 2008; Pabalan et al., 1998).

Model Validation and Predictive Sensitivity Analysis

Historical groundwater data have exhibited few detections of radionuclides. As such, a rigorous calibration exercise is not warranted or justifiable. However, the historical data will be used to validate the modeling calculations and potentially bound the values of some parameter combinations by simulating current conditions prior to undertaking predictive calculations. Multiple simulations will be conducted to evaluate the range of forecasts of possible impacts on groundwater beneath the landfill, at the property fence line/boundary, within surface water, at any defined receptors, and at any other locations of interest. Multiple scenarios will be simulated and predictive sensitivity analyses will be used to evaluate the potential impact of parameter variability on model outcomes at these locations. Although outside the scope of the proposed modeling task, the results of multiple-scenario and parameter-/prediction-sensitivity analyses can help guide the sampling frequency for long-term monitoring programs by providing a range of possible arrival-times and peak-concentrations for contamination at identified compliance locations such as the property fence line/boundary.

Commented [cao44]: Board noted MCL exceedances and their significance in Superfund program – see comment 15 above

Commented [cao45]: Gist of Board's spring 2012 draft recommendations/comments is that more data/wells are needed – so would so just using historical data address Board's recommendations/concerns?

Deliverables

The final deliverable anticipated to be developed from the modeling effort is a Technical Memorandum documenting the technical approach, assumptions, model development, parameterization, simulated scenarios, and results obtained. However, it is anticipated that there will be communication and interaction with USEPA to seek input on the FEPs, simulation scenarios, and parameter ranges and uncertainties identified for inclusion in the modeling prior to undertaking the model calculations. Communication and interaction with USEPA will include the following:

- Presentation and discussion of certain detailed or fundamental concepts – such as the CSM, FEPs and scenarios for inclusion in the modeling;
- Discussion of other less critical aspects of the modeling task; and
- Presentation of intermediate deliverables to USEPA for review and discussion.

No revisions to the SFS report are expected to be required as a result of this modeling effort.

Commented [cao46]: Is this premature conclusion?

Schedule

It is anticipated that the geochemical evaluation of potential leaching of radionuclides, including preparation and submittal of the Technical Memorandum, will be completed within twelve weeks of the approval to proceed.

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Commented [cao47]: what about EPA's rad and other CERCLA guidance documents?

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